SOME EARLY LEAD SALT DETECTOR DEVELOPMENTS*

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PREFACE

On 4 and 5 June 1930 a joint meeting of the Physical and
Optical Societies in England was called to discuss "Photo-Electric
Cells and Their Applications" (Anderson 1930). At that time,
the only photoelectric cells capable of a response in the infra-
red were selenium cells and the "thalofide" cell (Case 1930).
Experimenters intent on infrared investigations at that time
were otherwise constrained to use thermopiles or bolometers.
Such thermal detectors are too slow and insensitive for many
modern applications. The recent expansion of infrared science
and technology is largely based upon the availability of fast,
sensitive, and reliable cells.

Today, a wide variety of photoelectric cells are available
with responses to 1000μ and beyond (Smith 1965, Putley 1965).
The development of the first cells with a useful infrared re-
sponse is of interest. It involved scientists in nations
warring against one another. It involved scientists in univer-
sities, in industries, and in government laboratories. It in-
volved efforts ranging from technologic studies which provide
manufacturing capabilities to scientific investigations of
the basic processes involved.

Hopefully, the essays presented here describing a variety
of efforts to develop infrared sensitive lead salt cells will be
of some interest to a variety of readers. It is important to

young scientists to obtain some familiarity with the paths their predecessors took. The variety and diversity of activity in this narrow facet of science should be of interest in gaining an insight into scientific development. Science does not appear to proceed on some prescribable course. Rather, one perceives science to progress by odd and often unexpected contributions. Sometimes the progress is scored in a university - another time in an industrial laboratory - and again in government facilities. Sometimes, science is advanced by the genius of a single investigator - another time by the vision of an industrialist. The utility of science and technology to pursue war is often the stimulus to support scientific and technical endeavors. And, of course, the curious stimulus afforded by the inter-communication of results should not be minimized.

The pursuit of lead salt photodetectors incorporates all these features. It was, accordingly, with enthusiasm that I embarked on the research from which these essays evolved. It is with a good deal of humility that I finally conclude a year-long investigation. The work was sponsored by the Air Force to provide some case histories of research programs. The essays, by no means, comprise a total account of the research on lead salts prior to 1955. It is hoped, however, that sufficient data are presented to imbue the reader with the

magnitude and diversity of the developments. It is felt that the significant programs sponsored by U. S. military programs are described.

The bibliography presented at the end of these essays undoubtedly contains many omissions. However, it certainly typifies the nature of the effort. These essays, however, are not a result entirely of studying the material indicated in the bibliography. The real insight into the research was gained through numerous interviews. This provided a broader understanding of the work than otherwise possible. It is impossible to reference the material gained in this way, and errors in interpretation of these interviews or results expressed here are entirely my own.

I would like to express my gratitude to the following for patiently recounting experiences with me and in guiding me to source material:

Robert J. Cashman	James Humphrey
Paul J. Ovrebo	Edgar Kutzscher
Raymond H. McFee	R. A. Smith
G. William Mahlman	Norman Anderson
Henry Levinstein	David Jeffries
Frank Bennett	Ronald Newburgh
Wayne W. Scanlon	

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INTRODUCTION

The photoconductive effect was first reported in 1873 by
Willoughby Smith who had been experimenting with the use of
selenium as an insulator for submarine cables (Pearson and
Brattain 1955). He and an assistant noted that crystalline
selenium offers considerably less electrical resistance when
exposed to light than when it is kept in the dark. Since the
response of selenium extends slightly into the infrared, a
few scientists soon began applying cells made of this material
to a variety of investigations requiring an infrared sen-
sitivity (Pfund 1904). However, the cells were unreliable
and the measurements made in this manner had little impact on
science, nor did the desire to have such cells provide a
strong impetus to manufacture photoconductive cells.

By the mid-1930's, reliable photocells that utilized
photoemissive surfaces were being manufactured. Photovoltaic
and photoconductive cells on the other hand were laboratory
instruments primarily used for studies of their material be-
havior (Allen 1930). As World War II approached, the desire
to "see" and communicate unobtrusively goaded the military
services (both in Germany and in the U.S.A. and England) to

support efforts to develop improved photoconductors with a sensitivity in the infrared. Such cells have since permitted the development of numerous military devices for communication, detection and tracking, and for the homing of guided missiles. Fortunately, the cells have also been useful in the advance of astronomy, geology, medicine, agriculture, meteorology, and other scientific disciplines (Ballard 1959).

The first important photoconductive detector with a significant infrared response was lead sulfide. It has sufficient spectral sensitivity to permit its broad application to many military problems. With the successes obtained with this material, attention was soon given to other lead salts having a longer wavelength response.

The developments described are only loosely related. They were often practised under military security or under some degree of industrial proprietary regulation. One does note, however, that the pioneering work of Robert J. Cashman in the U.S.A. has influenced to some degree most of the programs to be described here.

Present day progress in detector developments are generally undertaken with good communication between groups. Of course, proprietary information when it exists or is thought to exist is jealously guarded. It is beyond the scope of these essays to

examine the causes of this change. The change is reflected,
however, in the Photoconductivity Conference held in Atlantic
City on 4, 5, and 6 November 1954 (Breckenridge, et al 1955).
Although several reports on photoconductivity reached the
open literature prior to this time, this conference marked a
rapid rise in reporting investigations on photoconductivity.
It is, accordingly, appropriate to end the essays presented
here at about that point. It will also be necessary, accord-
ingly, to consider each investigation individually rather
than chronologically.

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MILITARY OBJECTIVES

2.1. The U.S.A. Wartime Developments

The events in Eurpoe prior to 1940 revealed clearly to

Harvard President James Bryant Conant, MIT Vice President

Vannevar Bush, and others that American science must be

organized to meet the impending threat. Recommendations to

that effect, under Bush's leadership, were made to President

Franklin Roosevelt (Bush 1945). As a result, the National

Defense Research Committee (NDRC) was formed on 27 June 1940.

This was reorganized a year later when the Office of Scientific

Research and Development (OSRD) was established with NDRC as

one of its units. Pulitzer Prize winning author Theodore H.

White has asserted that although the hardware developed (atom

bombs, radar, sonar, antibiotics, etc.) changed history, the

"greatest invention was, perhaps, political: a new way of

using brains" (T. H. White 1967). The impact of the OSRD

programs certainly had a pronounced effect on the course of

history. It is the intent here to draw attention to a re-

latively small aspect of the total OSRD activity and to note

its impact on the history of technology.

Late in 1942, an Optics Division (Div. 16) of NDRC was

formed, largely as a reorganization of an older Instruments Section. George R. Harrison headed the activity of this group (Owens 1946). It carried out a broad program during the war years, including studies involving the principles of techniques of camouflage, improvements in aerial mapping and photography, the development of devices to detect aircraft by infrared radiation, techniques for intercommunication with infrared radiation, and the development of the sniperscope. These general developments have been ably described (Arnquist 1959), so that the discussion here will be directed toward programs requiring improved infrared radiation detectors. These were mainly undertaken within Section 16.4, which was charged with the development of non-image-forming infrared equipments.

O.S. Duffendack, who was chief of Section 16.4, led investigations in two spectral regions: the near infrared (from about 0.8 to 1.5μ) and the far infrared (from about 5 to 15μ). The near infrared utilized thallous sulfide detectors and were active systems (i.e. an emitting source was required). The far infrared utilized thermal detectors and passive systems (i.e. the self-emission of all heated objects was the source). Note that at present the terms "near" and "far" as then used are no longer relevant since infrared technology now extends to the millimeter spectral range, and this usage in describing infrared phenomena has virtually ceased.

The far-infrared devices were developed for: (a) the detection and location of personnel, vehicles, tanks, aircraft, and ships, (b) ranging (Strong 1945a), and (c) guidance of missiles. The effective range of these devices varied from a few hundred yards for personnel to about 12 miles for a ship in clear weather. The range was also drastically reduced by clouds and fog. Research was undertaken to evaluate various thermopiles, evaporated metal-strip bolometers, and various types of thermistor bolometers.

The need for improved infrared detectors to be applied in many of these systems was obvious, and detector research was undertaken from a variety of directions. Experimental and theoretical investigations of photoconductors were supported (von Hippel 1943, von Hippel 1945, von Hippel, et al 1945, Hole and Cashman 1945). Programs to improve thermal detectors were pursued (Harris 1945). Infrared sensitive phosphors were also studied (O'Brien 1946, Urbach, et al 1946). It is the intent here to concentrate attention on the developments of photoconductors.

The infrared characteristics of the emission from various military targets was studied (Butler 1944) as were the radiation characteristics of backgrounds (Strong 1945c) and the atmosphere attenuation of infrared radiation (Strong 1945b).

Wartime research in the U.S.A. on photoconductors proceeded initially with investigations of silicon cells (Teal 1944) and thallous sulfide cells (Cashman 1943, 1945a). The latter were found to be the better and were extensively applied to optical communication systems (Spence 1945, Lothrop, et al 1945, H. E. White, et al 1945, Snyder and Platt 1945, Fluke and Porter 1946), and to a "Japir" device (Geiger and Black 1945) which could be used to determine if the enemy was employing infrared signaling.

The thallous sulfide detector was limited in use to active systems, i.e., those utilizing an infrared irradiating source. However, the development of this detector was significant in that it permitted an understanding of the sensitization process which could then be applied to other materials (Lovell 1968), and particularly, lead sulfide. It is the intent of this section to emphasize the contributions of Robert J. Cashman in developing the lead sulfide cell.

The pioneering work Cashman did was, of course, in complete ignorance of developments in Germany which preceded his (see Sec. 2.2) Cashman's work, moreover, has had a strong influence on subsequent developments in this country.

As a youngster, Cashman made selenium cells and performed simple experiments with these. Partially for this reason he studied physics in college, receiving his bachelor's degree from

Bethany College in 1929. Transferring to Northwestern University, he obtained an A.M. degree in 1930, and a Ph.D. in 1935. His interest in devices which convert photon energy to electrical energy was enhanced by his formal training.

Cashman stayed on at Northwestern, joining the faculty. His research efforts continued in investigations of transducers of radiation to electricity. In particular, he studied the photoemissive properties of metals. By 1939, he began examining the photoemissive characteristics of semiconductors in hopes of finding a simpler mechanism, and thus of providing an improved understanding of semiconductor behavior. Among the semiconductors Cashman had studied were Tl_2S, Tl_2Se, Tl_2Te, MoS_2, Ag_2S, and PbS.

During these investigations, Cashman noted the photoconductive effects, but these were subsidiary to his main interests. In the U.S.A. no one, to Cashman's knowledge, was investigating photoconductive phenomena, although studies of semiconductors were well under way.

As the threat of war loomed, Cashman was asked by NDRC to turn his attention toward making reliable infrared sensitive cells, and on 1 December 1941, an OSRD contract was initiated which authorized Cashman to work toward that goal. The contract was initially for a six month period at a cost of

$3000. It was extended throughout the war years, but the total billing came to only $98,385.46.* Initially, the goal of the contract was to determine techniques to manufacture stable, sensitive thallous sulfide cells. When that technique was developed, Cashman was able to turn his attention to other materials.

The thallous sulfide cells were desired for use in an infrared communication system. During the first World War, T. W. Case had experimented with this concept. He had successfully sent messages over a distance of 18 miles, but his detectors were unreliable. In particular, they suffered fatigue when exposed to short wavelength radiation. Neither could cells be manufactured with reproducible characteristics. And with that war's end in 1918, interest in further development ceased (Case 1917, 1922).

Cashman found that he could evaporate Tl_2S over a grid on the inner surface of an evacuated tube. By diligent experimentation, he discovered techniques to provide optimum sensitivity to the cell. He soon perceived that the necessary ingredient was oxygen.

By late 1943, the developmental work on thallous sulfide

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*Contract OEMsr-235 file, NDRC Section 16.4, National Archives, Washington, D. C.

reached a point where cells of excellent characteristics could repetitively be made. This permitted the process to be turned over to consideration of large scale manufacturing techniques (Hewlett, et al 1945) and freed Cashman to investigate the properties of other materials.

An urgent need had arisen for photoconductive cells with a spectral response to longer wavelengths. Cashman began investigations of MoS_2 and Ag_2S, and in February 1944 he undertook an active program with PbS (Cashman 1945b). During the first half of 1944, a large number of natural crystals of molybdenite crystals gave the stronger response and, at that time, the expectation was that MoS_2 would prove to be the superior detector.

Synthetic photoconducting layers of Ag_2S, MoS_2, and PbS were also studied at that time. The first two materials were found to decompose at temperatures near their melting point, ruling out the possibility of deposition by evaporation. Chemical deposition was investigated, but responses were far lower than those obtained for thallous sulfide.

The investigations thus indicated that PbS had the greatest potential as an infrared detector. By September 1944, Cashman was able to show conclusively that oxygen photosensitizes lead sulfide. By December, cells of sufficient response had been

made to permit measurements of the spectral response and the time constant. These indicated a response to wavelengths longer than those to which Tl_2S responded, a shorter time constant, and a lower noise. However, the signal response was still below that of Tl_2S. Cashman thus directed his research efforts toward the development of PbS cells with greater responsivity.

Evaporated cells were made by placing about 10 milligrams of lead sulfide powder in a small tube. This was pumped down to a pressure of 200 microns, and a hand torch, using a gas-oxygen fuel was used to slowly heat the powder. The lead sulfide then evaporated and condensed on the wall of the tube on which electrical leads were drawn with aquadag.

By December 1944, Cashman was preparing chemical depositions of PbS, and his activation studies paralled those of his evaporated films. His chemical techniques were adopted from techniques well known to make "black mirrors." Dilute solutions of lead salt and thiourea decompose in the presence of an alkali, and lead sulfide is deposited on the container walls as well as on glass plates inserted into the solution for that purpose.

The chemically deposited layers always indicated some initial photoconductivity, although the effect differed markedly between cells. Since Cashman had conclusively shown that oxygen photosensitized evaporated layers of PbS, he assumed that oxygen (or

oxygen-containing compounds) must be responsible for the photo-sensitization of chemically deposited cells.

Most of the sentitization experiments were purely empirical, since very little in the way of a theoretical understanding could guide the investigations. Cashman subjected the chemically pre-pared layers to heat treatments in air and in oxygen at a variety of pressures, and in a vacuum. The signal response was found to increase somewhat for heat treatments in air at temperatures less than $125^{\circ}C$ and for periods up to 2 hours. Surprisingly, the best results in signal response were obtained by heat treat-ments in vacuum. These lasted up to 5 hours at temperatures not exceeding $150^{\circ}C$. However, an increase in cell resistance from about 0.1 to 10 megohms was also noted.

Heat treatments at temperatures above about $170^{\circ}C$ caused the layer to lose its photosensitivity permanently, and to suffer a change in color. By subjecting the layers to intermittent heat-ing with a gas-air flame in direct contact with the layer, an im-provement in photosensitivity resulted. This suggested to Cashman that compounds other than PbS are present in the layer, which would decompose at temperatures above about $170^{\circ}C$ and give rise to the loss of photosensitivity and change in color. The result obtained by the sudden heating further suggested that the

foreign material was driven out but that the time to reduce
PbS to an insensitive oxide was insufficient.

Cashman carefully measured the performance characteristics
of his cells. He noted a temperature coefficient of resistance
of about 2 per cent per degree centigrade, as compared to 6 per
cent for thallous sulfide cells. The ratio of the lead sulfide
cell maximum to minimum response in the region where the re-
sponse is linear with incident flux was found to be 6×10^6,
which is considerably better than that for thallous sulfide.
The spectral response observed for three cells is reproduced in
Fig. 1. He noted that the threshold in all cases appears to
be near 3.6μ, but the wavelength of peak sensitivity depends
upon the activation process and cell thickness. When the cell
was cooled to dry ice temperature $(-80^{\circ}C)$ the spectral response
changed, but in 1944-45, this could not be explained. The
noise characteristics of the cells were also measured, rather ex-
tensively. Cashman noted the effect of cell area, the effect of
applied potential, the frequency characteristics, and the effect
of background. This was a big stride toward elucidating the
mechanisms of photoconductivity.

In December 1944, Cashman was sent some captured German photo-
conductive cells, made by Zeiss Ikon (see Sec. 2.2). The sensitive
area of each was approximately 0.5 x 0.5 mm. The color of the cell
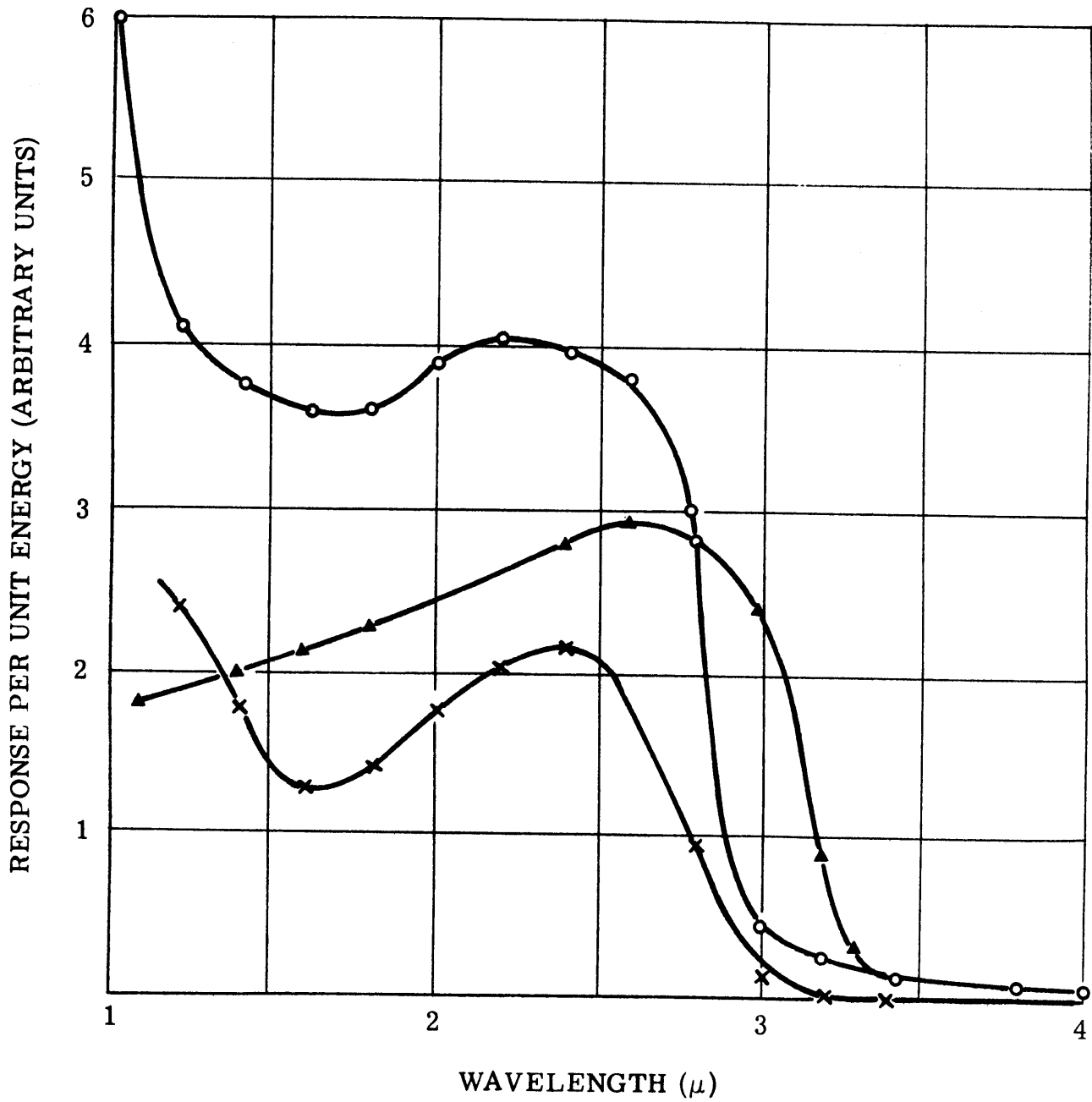
SPECTRAL RESPONSE CURVES OF THREE LEAD SULFIDE CELLS

(Cashman 1946b)

indicated it was PbS. Subsequent studies with x-ray diffraction and spectroscopic analysis also suggested it was PbS. Cashman's cells were not quite as sensitive, owing to their larger size. However, by the following February, Cashman had made cells with the same area as the German cells. His showed superior performance (Cashman 1947a).

The knowledge that the Germans possessed PbS cells goaded an interest in applying Cashman's cells in U.S.A. infrared systems. A group at the University of Michigan was particularly interested in applying them to locate military targets. Its tests indicated that PbS was considerably superior to Tl_2S in many applications. However, the war ended before the systems could be effectively changed, and the Michigan activity ended, reports could only refer to the promising characteristics of PbS with recommendations to pursue that development (Black, et al 1945).

In the winter of 1945, the British (see Sec. 2.4) were advised of Cashman's success with PbS. In March 1945, and again in August 1945, British scientists visited Cashman.

In November 1945, the NDRC contract terminated, but the work was continued under a Bureau of Ships contract. Demands for Cashman's cells grew as the word of Cashman's successes spread.

During the World War II years, the value of science in enhancing military success was firmly implanted in the minds of

military management. Consequently, support of some technical research and development programs continued. Programs requiring infrared sensitive detectors flourished because of the availability of reliable photoconductors.

Cashman himself has continued his photoconductor research, extending his work to all the lead salts (Clark, et al 1951, Cashman 1949, 1954, 1955, 1958, 1959, Cashman, et al 1951). His activity has influenced all subsequent U.S.A. investigations either directly or indirectly.

2.2. German Lead Salt Developments

Historically, the German investigations of lead salt detectors significantly antedate those undertaken in the U.S.A. However, their impact on U.S.A. investigations was felt after the independent discoveries of Cashman. This, naturally, was due to the German military restrictions imposed on disseminating information regarding their work. On the other hand, British investigations of lead salt detectors followed the discovery by British intelligence of the German activity (see Sec. 2.4).

One of the German pioneers was Edgar W. Kutzscher, who in 1930 began investigations at the Department of Physics at the University of Berlin of the electrical and physical characteristics of crystal rectifiers (Kutzscher 1949). He soon discovered that some of these crystals generated an electromotive force and/or changed

their resistance when exposed to visible or infrared radiation in the vicinity of the "cat whisker" electrode used to touch the crystal. Kutzscher was aware that other investigators had also noted similar effects, but no comprehensive investigations had been undertaken.

Kutzscher then began investigations to determine what materials exhibit these photoelectric effects; the relationship (if any) of the rectifying and photoelectric properties; the total sensitivity, spectral sensitivity, and time response of the cells; the relationship between the electrical response and the distance between the irradiating point and the metalic contact to the crystal; the effect of the shape of the contact; possible means to improve the sensitivity of a crystal; and possible means for shifting the spectral response.

In the course of these investigations, Kutzscher and a collaborator discovered that galena (PbS) crystals could be sensitized to have a short wavelength infrared sensitivity superior to that of any other radiation detector then known. The German military agencies took an interest in these investigations since they were interested in communication devices employing infrared radiation.

Beginning in about 1933, Kutzscher's work was supported by the German Army. The military also was interested in locating

ships, and it was soon found that PbS cells as receivers gave results comparable to those of the bolometers manufactured by Zeiss for that purpose. Attention was then given to using PbS cells in devices to detect aircraft, tanks, and other military targets.

The original hope for the utilization of infrared sensors had been to assist radar. About 1936, radar devices had too wide a field to precisely locate an object angularly. This an infrared device can do, but of course no range information is attainable. When radar was improved to permit angular resolution, interest in infrared faded. However, the realization that radar is susceptible to jamming caused a renewed interest in infrared.

The early cells utilized the "cat whisker" electrode and showed a sensitivity only in the immediate vicinity of the electrode. In order to improve the performance of the cell, several small crystals were formed and connected in series to comprise a single cell. Each crystal had its own contact, and such a cell with a large sensitive area provided improved performance, but the fabrication required great skill and considerable time.

Kutzscher's investigations led to the preparation of PbS films which could be used as photoconductive cells. Both

chemically precipitated and evaporated depositions were studied.
The chemical precipitation method used as basic materials lead
acetate, sulfur carbamide (thiourea), and sodium hydroxide.
These three components were joined to obtain a precipitation of
lead sulfide on a glass substrate. It was then heated to pro-
vide sensitization. The chemical process, the layer thickness,
the impurity content, and the subsequent heating were carefully
studied to determine their effect on the resulting cell per-
formance.

The German military services supported research and develop-
ment programs in some universities and private industries. Of
the latter the Electroacustic Company in Kiel was undertaking
the development of underwater sound equipment and the acoustical
detection of aircraft. Upon hearing of Kutzscher's successes,
they realized the potential of infrared devices and in 1937 hired
him to lead a development and production effort in infrared de-
vices. Kutzscher, by also maintaining his position as Director
of Infrared Research and Development at the Department of Applied
Physics at the Institute of Technology at Berlin thus had an
excellent ventage to direct the development of cells having optimum
characteristics for use in military devices.

It was soon found that the signal-to-noise ratio improved when
precipitated cells were cooled. Cells were thus formed on Dewar
flasks to permit cooling to liquid air temperatures. Since

cooling with liquids is cumbersome for field use, cells were soon made which gave satisfactory performance when cooled to dry ice temperatures (-80°C).

By 1944, approximately 600 men were working on infrared devices at the Electroacustic Company and 1000 cells were being fabricated annually. Of these employees, about 100 were involved in research and development activities.

In addition to Kutzscher's work on chemically deposited PbS cells, the Germans supported research on evaporated films (Gudden at the University of Prague) and on crystals (Görlich at Zeiss Ikon in Dresden). Good interchange of information was maintained between these groups. The resulting cooperation enabled the programs to proceed efficiently. It is interesting to note that on 2 and 3 October 1942, a meeting was held in Berlin of scientists involved in infrared research. The proceedings of this meeting were published.*

It was early recognized that oxygen plays an important role in sensitizing cells and in shifting the peak sensitivity toward shorter wavelengths. Since it was desired to obtain cells with longer wavelength sensitivity and thus to provide sensitivity to cooler targets, experiments were undertaken to determine if

*Forschungsarbeiten über ultrarotempfindliche Strahlungsempfanger, 2 and 3 Oktober 1942, Berlin.

other chemical elements in the oxygen group of the periodic system would provide such sensitivity. Accordingly, selenium and tellurium were used experimentally. The studies were of cells in which Se and Te were introduced as impurities in PbS layers and cells in which PbSe and PbTe supplanted the PbS. Cells produced in this manner were found to have the longer wavelength response sought, but their full potential could not be realized by the end of the war.

The development of lead salt detectors in Germany required some luck and much effort. The basic theory of semiconductor behavior was not known. All one could do was to investigate the effects of a single parameter; but often it is impossible to control the process so precisely that only one parameter varies. The recipes used for manufacturing cells thus were developed empirically. The concentrations of the chemicals, the temperature of the bath, the time of substrate introduction, the stirring rate, and other aspects of the process resulted somewhat as an art.

Despite the empirical nature of cell manufacture, the Germans had managed to develop for military use several operational devices which respond to infrared energy. These involved ground-based devices for detecting and tracking aircraft, and others performing a similar function on ships. Shipborne devices with

similar objectives were under development. Airborne devices to detect other aircraft, ships, and ground targets were also being developed.[*] Infrared devices were used to provide homing and a proximity fuse for guided missiles.

Realizing that the Kiel site was vulnerable since it was so close to the North Sea, the Infrared Branch of the Electroacustic Company was moved to Namslau in eastern Germany late in the war. In January 1945, fears that the Russians would capture this area prompted the Germans to plan an evacuation of the laboratory and manufacturing facility. However, the Russian advance was too fast to permit an execution of the plan. Hence, on 25 January 1945 the Russians occupied the factory and obtained possession of most of the records and equipment, thus providing the basis for their present infrared technology.

The following month Kutzscher tried to reestablish his group in Kiel. By April, Kutzscher was picked up by English agents and taken to London for interrogation. In 1947, he was brought to the U.S.A. where he continued lead salt research under U. S. Navy sponsorship.

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[*]One such device, the Kiel IV, was in the early stages of production in Jena when the allies overtook the area. This is discussed in Sec. 2.3.

2.3.   The Postwar Basis for Sponsoring Detector Developments by

the U. S. Military

As the military forces were being demobilized at the end
of World War II, a change in emphasis regarding research and
development programs occurred in the United States.   The intensive
efforts expended during the war years were replaced by a some-
what lethargic attitude.   At the same time, much of the security
which hindered information exchange was lifted and many new
researchers became interested in photoconductive effects.

With the disestablishment of OSRD, the individual services
continued to sponsor some research programs.   We have noted, for
instance, that the work of Cashman was continued under a Navy
contract.   Such programs were coordinated by the Research and
Development Board to assure that no duplication in effort took
place.   After this board was dissolved by President Eisenhower,
however, scientific progress accelerated as a result of the
improved cross-fertilization of ideas.

It was not until 11 August 1949 that the Office of Naval
Research Branch Office, Pasadena, began sponsoring joint-service
classified symposia devoted to the military applications of
infrared radiation (Laufer 1957).   This was the forerunner of
the organization now called the Infrared Information Symposia
(IRIS).   This marked the beginning of interchange of information,

but as late as 1951 many Navy officials still had strong doubts as to the ability of infrared techniques to solve military problems. A summer study, called Project METCALF, was held that year to establish firm thresholds of performance that could be expected (Isakson 1966).

The situation in the Army was similar. It was characterized by the considerations given at the Air Materiel Command, Engineering Division,[*] located at Wright Field, Ohio, to captured German infrared equipment and the subsequent development there of similar equipment. Those considerations tend to typify the R & D attitudes of the time.

In April 1946, a Kiel IV device was brought to Wright Field for extensive airborne tests. The Kiel IV was an infrared sensitive seeker which the Germans had begun to put into production as the war ended. The American forces had found the equipment in an assembly line at the Carl Zeiss plant in Jena. The parts were shipped to Fort Belvoir, where the assembly was completed. Werner Weihe, who had supervised the Jena effort, was then brought from Germany to supervise the Belvoir studies.

The Kiel IV was intended for use in aircraft to detect other aircraft at night. It utilized a PbS detector manufactured by the Electroacustic Company in Kiel and the output was displayed

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[*]Note that the Air Force was not established until 18 September 1947.

on a cathode ray tube, permitting the pilot to home in on a target. The Corps of Engineers at Fort Belvoir tested the instrument, and concluded that "... although lead sulfide cells have been made in this country, the Germans were far ahead, both in the knowledge and in the manufacture of these cells." They also heralded lead selenide cells and forecast lead telluride, saying "it is expected that in the near future advances will be made in the field of photo semiconductor cells. It is probable that more sensitive cells with response to longer wavelengths will be developed" (Meservey and Taylor 1946).

They recommended that "... research be continued and expanded on infrared photoresistors with the objectives of increased sensitivity, decreased response time, and increased response to longer wavelengths." It was also at their recommendation that Weihe and the instrument were brought to Wright Field.

The insouciance associated with these tests has been described (Neal 1956). Sgt. Theodore Fields, who aided Weihe, went on furlough from 20 May until 10 June in the midst of the period planned for testing. The infrared activity was moved from one building to another in this period and at least another week was lost. Paul J. Ovrebo, who was in charge of the infrared activity at the Engineering Division, was sent out recruiting for a month and thus was not available to participate.

Nevertheless, the Kiel IV was mounted on a C-45 and tests were run between 16 and 23 July 1946 with a B-17 as a target. Both Ovrebo and Fields took part in the tests. Ovrebo had left the Physics Department at Susquehanna University in 1943 to join the Signal Corps in Dayton, Ohio, remaining through the re-organization of the Engineering Division. The experience with the Kiel IV incited his interest in infrared, and he remained at Wright Field for several years to head much of the important development work sponsored there.

The July tests of the Kiel IV indicated that it had a useful range of 4.3 miles at an altitude of 1 mile, which was extended to 6.8 miles at an altitude of 2.5 miles. The Army Air Corps planned to extend those tests by: (a) installing the set next to a radar to get exact ranges, (b) testing against ground targets such as steel mills and refining plants, and (c) testing against other airborne targets such as jets. However, on 22 July 1946 the Corps of Engineers demanded the return of the equipment "... in order that other planned work may take place without further delay."*

The plans for further testing at Wright Field were, therefore, canceled. It is interesting to note that Neal quotes Ovrebo as being convinced that the returned equipment was "... promptly and

*Air Materiel Command, Engineering Division, Memorandum Report, "German Infra-Red Airborne Detector Kiel IV", 30 Jan. 1947, Air Force Archives.

permanently placed on a shelf."

The well-known ability of a soldier to scrounge, however, permitted Wright Field to continue the investigations. Upon returning from a visit to Fort Belvoir, Sgt. Fields had possession of the scanning parts of the Kiel IV. The Navy was able to supply some electronic parts. Heinz Fischer, who had come from Germany to Wright Field, recruited George Klinger, also a German, to join him in January 1947 to reconstruct an operational Kiel IV. Klinger, with no knowledge of English, worked steadily and by 5 April 1948 had an infrared search set ready for flight tests. This instrument was given the nomenclature AN/AAS-1 (XA-1).

These tests indicated the need for further modifications, and by 5 October 1949 the XA-2 was available. All of these instruments were capable only of looking ahead or slightly up, and were thus useful only in air-to-air missions.

On 7 September 1950, an operations analyst with the Tactical Air Command in Korea wrote to Ovrebo to enquire if "... the German device on which you have been working for some time might be adopted for use not only in detecting ... night movements of troops, tanks, etc. ... but also in directing gun fire, rockets, or bomb drops to stop such movement."[*]

The tests that had been performed were insufficient to provide

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[*]Letter from Carroll W. Bryant, TAC Operations Analyst to Paul J. Ovrebo, Wright Field, 7 September 1950, Air Force Archives.

a satisfactory answer. Modifications were immediately begun to permit looking down with the equipment and tests undertaken to indicate its potential. Tests undertaken during the winter of 1950-51 convinced Ovrebo that a power plant could be detected at a slant range of 10 miles (Ovrebo, et al 1951).

In the spring of 1951, Luther Meuser and DeMetro Cavitch demonstrated the XA-2 at the Aberdeen Proving Grounds, Maryland. Following this, the Tactical Air Command (TAC) in Korea informed Washington of a requirement for infrared detection equipment in Korea. The TAC requested six production models to be built within six months. By June 1952, aircraft with the new XA-3 equipment arrived in Japan. The first infrared combat mission in Air Force history was recorded on 18 September 1953.

Although infrared-sensitive instrumentation had by this time become operational for the military forces, the incentive to undertake PbS cell development had by no means been fulfilled. On the contrary, on one occasion during the Korean conflict, only two working PbS cells were available in Japan for the XA-3 equipment. The lesson was clear that support of both research and development of photoconductive devices had to be maintained.

2.4. Lead Salt Research in Great Britain

We have noted that although the German developments of PbS cells preceeded the American by about a decade, Cashman's work

before December 1944 was done with no knowledge of their successes. However, it was the discovery by British intelligence of the German successes (Kutzscher and his associates at the Electro-acustic firm in Kiel) that prompted British investigations of lead salt detectors (Elliot 1947). The initial activity was undertaken at the Admiralty Research Laboratory. However, quite rapidly studies were undertaken in other laboratories, principally the Telecommunications Research Establishment (now the Royal Radar Establishment) and at the Cavendish Laboratory at Cambridge.

Characteristic of the British, much of their effort was directed toward an understanding of the basic phenomena under-lying the photoconductive mechanism. However, one must not assume that their interest in lead salt detectors was merely academic. On the contrary, extensive efforts were made to detect the hot gases emitted by schnorkelling submarines. They also used lead salt detectors in guided missiles to attack the hot jet pipe of jet aircraft. As a result, the Mullard Company established commercial production facilities, initially of PbTe. This was later abandoned in favor of InSb detectors.

By 1948, several papers described the findings of these research investigations (Starkiewicz 1948, Starkiewicz, et al 1946, Sosnowski, et al 1947, Sosnowski, et al 1947, Moss and Chasmar 1948, Simpson 1947, Simpson 1948, Kicinski 1948).

Shortly thereafter, tutorial papers appeared describing
British achievements at length (Sutherland and Lee 1948,
Simpson and Sutherland 1951, Smith 1950, Smith 1951, Moss 1952).
One notes that the British were quick to study the characteristics
of all lead salts. They were also, apparently, the first to
develop bubble windows. These, because of their thinness,
transmitted well to nearly 7μ. This technique was adopted at
NOL and is described in Sec. 5 of this report.

The work of the British by 1950 was in the forefront of
our knowledge. The number of workers also expanded (Braithe-
waite 1951, Roberts and Young 1953). Hand produced cells of
exceptional quality were also made available to enable in-
vestigators to carry out high resolution spectroscopy to a
degree not previously achieved. E. K. Plyler at the National
Bureau of Standards received a PbTe detector from RRE, as did
H. W. Thompson at Oxford. The impact on American investigations
was further enhanced by frequent visits as well as the temporary
acceptance of British scientists as professors of physics at
the University of Michigan (Simpson and Sutherland 1952).

The essays presented here consider primarily the first
decade following Cashman's initial discoveries. The British
developments in this period are summarized quite extensively
(Smith 1953, 1956). In these summaries, it is noted that the
main characteristics to be observed are the width of the energy

gap between the full and conduction bands and the nature, density, and distribution of impurities as a function of temperature. The mobility of electron and hole carriers and their effective mass must also be ascertained. It was also pointed out that measurements made on films are quite limited so little progress was made until pure crystals could be grown. The measurements made were reviewed along with a description in some detail of a theoretical interpretation of them. It is unnecessary to further describe these summaries. Note, however, that American investigators in the mid 1950s had access to the reports of the British, as well as occassional contact.

3

THE ECA PbS INVESTIGATIONS

The Electronics Corporation of America (ECA) was founded in 1937 under the name Photoswitch, Incorporated by Arthur G. B. Metcalf to supply electronic control systems. These products found extensive industrial applications and were also adaptable to war needs during World War II. The company is thus somewhat typical of the scientific-military-industrial complex which grew in this period (Nieburg 1966). Blossoming on the periphery of a great university (M.I.T.), it was stimulated by contact with faculty members, and the student body provided a reservior of employable talent. Both Wayne B. Nottingham and John C. Slater, M.I.T. professors, played important roles in ECA's PbS investigations.

Metcalf was well known at Wright Field, and in the summer of 1946 visited Faye E. Null there. Null at that time was a civilian scientist with the Army Air Corps. He, being familiar with the remarkable successes of Cashman and others working on OSRD projects, was stirred by the possibilities of "seeing" in the infrared. He envisaged a mosaic tube of PbS cells which would develop images much as a television tube does. Metcalf became interested and, anxious to expand his electronics

research activity, proposed a program directed toward fulfilling
Null's vision.*

On 16 September 1946, a contract for $40,000 was let to ECA
to "... conduct a study covering fundamental research in the field
of semiconductors, with the ultimate aim of obtaining improved
sensitivity within the ranges of all atmospheric windows within
the infrared region of the electromagnetic spectrum. Specifically
included in the research and forming the initial ground-work
of the investigation, will be the production of improved PbS
cells and mosaics."**

It must be reemphasized that in 1946 hardly anyone in the
U.S.A. but Cashman and those who had worked with him had ex-
perience with research in photoconductive detectors. The first
task facing ECA thus was to build a technical staff. In November
1946, Raymond H. McFee, who had received his Ph.D. at M.I.T. in
1943, was hired to direct the research effort. He had little
more to direct this with than the reports of Cashman that he could
secure.

Although there was precious little experience to rely on,
the potential of infrared measurements possible with improved

*Letter from A.G.B. Metcalf to Headquarters, Air Materiel Command,
Wright Field, Dayton, Ohio, Attn: TSBPE-7B, 20 August 1946, Air
Force Archives.

**Contract No. (33-038) 47-651, Air Force Archives.

detectors provided a powerful incentive. ECA secured the con-
sulting assistance of men prominent in investigations of semi-
conductors. Nottingham and Slater of M.I.T. were both involved
in the ECA investigations. In addition, Metcalf secured the
consulting services of Karl Lark-Horowitz of Purdue. Since
these men were scientists of the highest reputations, arranging
their consulting services presented considerable problems. ECA
also planned to secure the consulting services of Cashman. How-
ever, security restrictions complicated this. The Navy was slow
in releasing Cashman. Nottingham was installed as chairman of
their consulting board.

At this time Col. Fred R. Dent, of the Air Materiel Command,
noted that Cashman had not secured a consulting arrangement with
ECA and that the contract had been "... negotiated on a sole
source basis, with the availability of Dr. Cashman as justification."[*]
However, the situation was explained to the satisfaction of Col.
Dent.[**] Efforts were continued to hire Cashman as a consultant,[***]

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[*]Letter, Col. Fred R. Dent, A.M.C. Eng. Div. to Photoswitch, Inc.,
5 November 1946. Air Force Archives.

Letter, Col. Fred R. Dent, A.M.C. Eng. Div. to A.G.B. Metcalf,
Photoswitch, Inc., 18 November 1946, Air Force Archives.

[**]Letter, Col. Fred R. Dent, A.M.C. Eng. Div. to Photoswitch, Inc.,
25 November 1946, Air Force Archives.

[***]Letter, Burton Shaw of Photoswitch to Robert J. Cashman, North-
western University, 13 November 1946, Air Force Archives.

and McFee visited him to learn experimental techniques. Notting-
ham and McFee soon had established an experimental program which
extended Cashman's process. McFee conceived of a modification
of this process whereby he improved the control. Like Cashman,
McFee painted aquadag leads on the interior of a glass cylinder,
and evaporated PbS in a partial vacuum (usually in a few tenths
of a millimeter pressure of oxygen) which condensed over the
leads to form the sensitive cell. However, McFee replaced
Cashman's torch with an oven to effect evaporation. By directing
a stream of cool air over the aquadag grid, he enabled the con-
densation to be localized.

A progress letter in November 1946 from ECA confidently
predicted the success of its process of "... a radically new
method of production which we have hopes will give us more
sensitive cells than have heretofore been produced."[*] It also
requested more classified reports, including Cashman's and
noted that the Navy had still not acted on releasing Cashman's
clearance to premit him to consult. However, the successes
that ECA had achieved permitted them to add, "... such consultation
is not a necessity with respect to the satisfactory conclusion
of this contract."

[*]Progress Letter No. 2 from Photoswitch, Incorporated to the
Air Material Command, 25 November 1946 Air Force Archives.

We have noted that in this period the military had not established clear cut objectives. Because of this uncertainty in objective, the Army decided not to continue this research in the Spring of 1947.* However, a month later, this decision was reversed, funds were obtained, and the program was continued.** This program then operated without interuption for 10 years, providing significant information and developmental techniques regarding the photoconductive exploitation of PbS.

The ECA staff engaged in PbS research gradually expanded. One of the first significant additions was Norman Anderson, who joined ECA in December 1948. He had obtained a Master's degree under Cashman during the war years, and had then gone to Continental Electric to supervise the manufacture of PbS cells there. These were expected to find a large market in sound motion picture pro-jectors. Because of their small size they could be readily pack-aged in a projector. However, with the advent at this time of safety film which is opaque in the infrared, the technique was no longer feasible. The experience had provided Anderson, how-ever, with insights into manufacturing PbS cells, which he applied at Photoswitch.

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*Letter of Col. J. C. Harvell, A.M.C., to Photoswitch, Inc., 9 April 1947 Air Force Archives.

**Letter of Col. J. C. Harvell, A.M.C., to Photoswitch, Inc., 2 May 1947 Air Force Archives.

Anderson's method utilized a cylinder on which PbS had been chemically deposited. The cylinder could be heated by a filament introduced on its axis. This assembly was then mounted on the base plate of a vacuum system. A second glass cylinder, on which an appropriate aquadag grid had been painted, was then placed coaxially over the PbS cylinder-filament assembly on the base plate. It was possible to evaporate the PbS, bleeding in O_2 for sensitization, and condensing the vapor on the outer cylinder to form a cell. This process led to improved uniformity and reproducibility, but the process still did not lend itself favorably to large scale production.

Consideration was given to chemical depositions. This, of course, would be more adaptable to commercial production. However, the sensitization required subsequent baking in air or an oxygen atmosphere at high temperature ($400 - 500^{\circ}C$). Ronald G. Newburgh, who joined ECA after graduating from Harvard, considered the problem and concluded that by using a chemical oxidant films could be chemically deposited which, by subsequent baking at a low temperature would achieve the desired sensitization. By this process, the oxidation is done in the interior of the film rather than on the surface. Further, reaction with the atmosphere is not required. The films produced in this way made possible an excellent yield with good uniformity and sensitivity. If high

temperature baking is used, recrystallization occurs which leads to added noise. The low temperature baking thus leads to cells with reduced noise levels. This process is the basis for most manufacturing of PbS cells today.

With the availability of good cells, studies of their fundamental characteristics were accelerated. These studies were based upon the operating parameters of the cells, such as spectral response, time constant, and noise characteristics. These extensive investigations were undertaken for two reasons: "In the first place, this understanding leads to the development of photoconductors with improved characteristics, since it shows specifically the line of investigation which should be followed in order to evaluate the photoconductors currently in production in a manner that allows for the quantitative evaluation of the influences of processing parameters. Secondly, the complete understanding of lead sulfide will show the way toward the better understanding of the lead selenide and the lead telluride photo-conductors as well as open the whole field of the creation of photoconductors with properties tailor-made to fit specific applications."[*]

The experimental investigations of cell characteristics was

[*]Photoswitch Technical Report 54-1, July 1954.

largely done by G. W. Mahlman, and the theoretical interpretations were done by Slater. Their results were reported fully in ECA technical reports to the sponsor as well as in the open literature (Mahlman 1953, 1956, Mahlman, et al 1956, Slater 1956).

4

THE EASTMAN PROCESS

The motivation which drew ECA into investigations of the photoconductive effects of lead sulfide was, as we have seen, deliberately based upon military requirements and the expectations that detectors could be developed having characteristics superior to the PbS cells then available. The situation at Eastman Kodak Company was a curious contrast to this.

In the late summer of 1947, Eastman was involved in manufacturing the Dove missile for the U. S. Navy. This missile used a cruciform shaped thermistor bolometer to sense the infrared emissions of the target it sought. However, Eastman wished to use an annular shaped bolometer, which it could not obtain from a supplier. The company, therefore, decided to investigate techniques to prepare its own. Eastman attempted to prepare sintered titanates as a thermistor material, but the process required high temperature furnaces and other manufacturing equipment not then available.

In December 1947, G. W. Hammar, in charge of the optics section of the Dove project, suggested the use of the semiconductor lead sulfide as a candidate for bolometer material. By mid-December, Frank Bennett, as Eastman chemist, had chemically deposited PbS from a reaction of a lead salt, sodium hydroxide, and

thiourea. Gold leads were deposited by evaporation, and the resulting cell was tested for its bolometric effect. This test involves determining the resistance of the cell as a function of temperature. Anomalies in the observations were soon discovered. These, in turn, were found to be related to turning on room lights, opening doors, or otherwise illuminating the cell. The cells were thus noted to have a photoconductive effect much greater than the bolometric effect!

Could such a detector be used as terminal guidance for an air-to-air missile? The cell was placed in a Dove sensing head and used to respond to a 100°C target. Significant signals were obtained, indicating an appreciable infrared response. With Navy approval, Eastman began to develop PbS photoconductive cells. E. D. McAlister, at Eastman, recognized the potential of infrared sensitive photoconductive cells, and was influential in the direction and support given to the PbS development program.

One of the first obstacles faced was the duplication of the photosensitivity accidentally displayed by the first cell. Everyone was aware, as a result of Cashman's work, that the presence of lead oxide in the film has a bearing on the photosensitivity. Attempts were, therefore, made to control the amount of oxygen present in the solution. The deposition was made in a highly alkaline solution. Since Courtney Q. Glassey, who temporarily

replaced Bennett as project chemist, recognized that both the sulfite ion and the metabisulfite ion are good removers of oxygen, he tried additions of sodium sulfite or sodium bisulfite to the solution. This had a twofold effect: (a) it provided a greater control over the rate at which the reaction proceeded, and (b) it served as a stabilizer for the thiourea, which had a tendency to deteriorate in time once it was put into solution.

Shortly thereafter, Glassey also demonstrated that the highest degree of photosensitivity produced directly out of solution came about by delaying the addition of the substrate to the reacting hydroxide-thiourea mixture. He noted, "... when thiourea is added to a highly alkaline lead salt solution, a glass plate or other material placed in the solution during the first minute or so automatically seeds or becomes seeded; that is, during the first minute or so there are formed on the surface centers or seeds at which the precipitated crystals can then form. However, after the first minute or so this automatic seeding normally terminates and if a plate that does not already contain its own seeds, is then placed in this solution such plate does not attract crystals of the precipitate (Glassey 1960)."

Glassey found it preferable to delay immersing the plate to be coated for at least five minutes to avoid seeding by the solution used for the final precipitation. It was, therefore,

necessary for the plate to be preseeded by another solution or process. The cells produced in this manner were thought to have a reproducible sensitivity which was 10 to 100 times better than theretofore achieved.

The seeding process was attributable largely to Meyer Sugarman. He described it thus: "... a glass plate is cleaned thoroughly and is then brought into contact with a special seeding solution either by dipping in the solution or having the solution poured over the glass plate....the seeding solution is made by adding to an aqueous solution of a soluble lead salt and a stabilizer (such as lead nitrate and polyvinyl alcohol) a solution of hydrogen sulfide in which the amount of hydrogen sulfide is more than necessary to convert all of the lead salt to lead sulfide (Sugarman 1959, 1960)."

Although Glassey designed the experiments that showed delayed action was advantageous, Hammar was probably the first to recognize the significance of the results and Bennett exploited the process and defined the limits. Many seeding media were tried before the PbS solution described. Of these, several gave good coats but poisoned the films so that no sensitivity developed.

Hammar and Bennett also found a technique to control the rate of precipitation. In this, "... the rate of precipitation of the lead sulfide and hence the crystalline form of the lead sulfide

coating is controlled by having a copper salt in the precipitation solution (Hammar and Bennett 1960)." The use of copper came in late 1948 or early 1949 after workers at Bell Telephone Laboratories had reported the effects of copper impurities in germanium transistors. Hammar and Bennett then made the assumption that contamination by copper was mitigating against high photosensitivity in PbS films. Rather than attempting to stringently eliminate all traces of copper contamination, Bennett decided to purposely contaminate lead sulfide with copper in a range of concentrations from 10^{-6} molar to 10^{-2} molar in solution. It was noted that copper had a decided effect on the rate of reaction, and if added at the proper concentration, whether by addition to the film or by some rate-controlling mechanism (which was unknown at the time), the addition of copper did give an enhancement of photosensitivity which was almost an order of magnitude over that which had previously been achieved.

The feature of the Eastman process was that the film as it came from the solution had a high degree of photosensitivity and did not require subsequent baking or oxidation. Further, it was stable in air, thus requiring no cover or evacuated envelope. In fact, the cells were unstable in a vacuum.

The Eastman process was essentially developed in secret. There was little interaction between Eastman scientists and those outside

the company, until late in the 50s (Spencer 1959). There were few contributions from Eastman in the open literature regarding PbS. The progress reports submitted to the Navy received little circulation. However, the Navy did receive good cells for military use – and thus the objectives of the program were met. Note, moreover, that technological developments are seldom published.

5

THE NOL PROGRAM

The potential of lead sulfide cells in applications of interest to Naval requirements was early recognized. In 1948, Lloyd Mundie at the Naval Ordnance Laboratory (NOL) began studying the properties of cells made by various people. This study compared the spectral sensitivity, the response to 200°C blackbody radiation, the time constant, and the variation of sensitivity over the surface of each cell available (Mundie, et al 1948).

Only 19 cells were available to Mundie for this investigation. However, it is interesting to note that even at this early date, he was able to obtain cells of German and English origin in addition to domestic units. The U. S. cells were produced by Cashman, Farrand Optical Company, Armour Research Laboratory, and the Radio Corporation of America. Neither Photoswitch (ECA) nor Eastman cells were yet available.

This was one of the first comparisons of the performance characteristics of a large number of cells with varied origin, and it did much to interest the Navy in the potential of infrared devices using photoconductive cells. Of particular significance is that it initiated a continued interest in research at NOL on the characteristics of photoconductive properties of the lead salts (Petritz and Humphrey 1958).

In 1948, Wayne W. Scanlon joined the NOL group. He had just
earned his Ph.D. at Purdue University under Lark-Horowitz, doing
his dissertation of the electrical properties of semiconductors.
At NOL, his first task was to review the existing literature on
photoconductive cells. In this he read an account of an inter-
rogation of a German scientist which indicated that PbTe might
make a cell with a response to longer wavelengths than that to
which PbS responded. Scanlon was quick to prepare a PbTe cell
by reacting stiochiometric proportions of Pb and Te, and
evaporating this in an evacuated pyrex container in which suitable
aquadag electrodes had been painted. These crude cells were sen-
sitive to infrared radiation when cooled to liquid nitrogen
temperature, but the absorption of the pyrex envelope limited
their spectral sensitivity to about 3.5μ. Very likely, these
were the first PbTe cells made in the U.S.A.

Scanlon soon learned of a technique used by Simpson in England
to make thin bubble windows which decreased the absorption beyond
3.5μ, permitting useful responses to 6μ. A year later (1949),
Simpson visited NOL and together with the NOL glass-blower Wellman
L. Clark, additional improvements on the technique were accomplished.
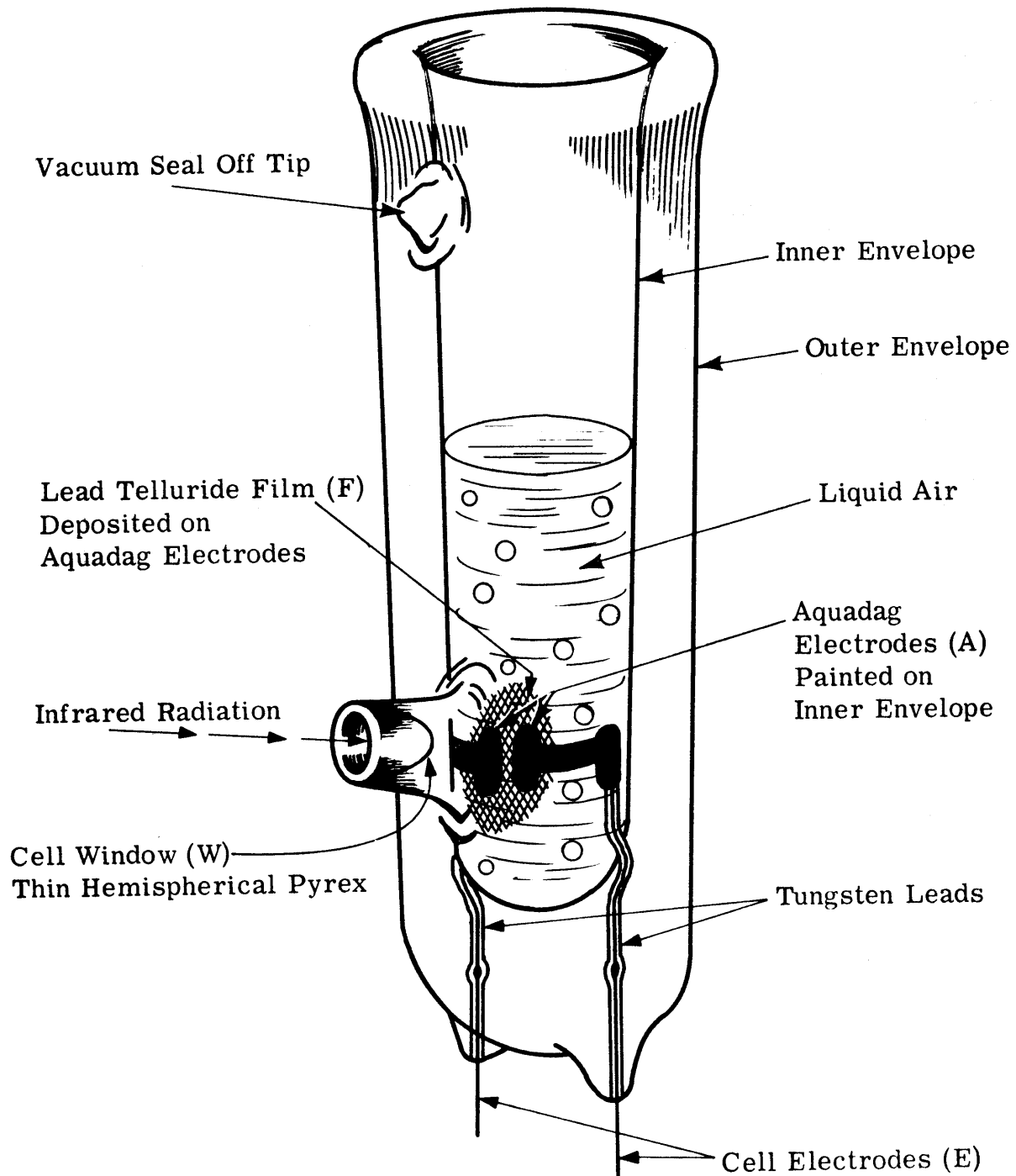
It is curious to note that the activation of these cells was
probably attributable to the meager facilities available to the
NOL scientists at that time. They were then quartered in the
Naval Gun Factory in Washington (and have since moved to facilities

at White Oak near Silver Spring, Maryland). The vacuum system then available lacked adequate performance characteristics to achieve a good vacuum. Consequently, the partial residual pressure probably provided the necessary oxygen for sensitization. Whatever the cause, usable cells were prepared and the performance characteristics could be measured (Scanlon 1951).

The cells were made from extremely purified lead telluride, since even minute traces of impurities seriously affect the photoconductive behavior. Lead was obtained from the American Smelting and Refining Company which was 99.999% pure. However, commercially available tellurium contained large impurity traces. A tellurium purification process was undertaken in which the impurities were removed by sublimation.

The PbTe cells made were found to be essentially insensitive at room temperature, so that provisions for cooling the films with liquid air were required. The films also became insensitive when exposed to air at atmospheric pressure. A cell thus was made as shown in Fig. 2.

With these successes in preparing usable cells, Scanlon chose to return to research in the semiconductor properties of the lead salts. He obtained galena crystals from the Smithsonian Museum in Washington. From these he obtained measurements of the forbidden band gap, mobility, mean free path, and other fundamental characteristics of PbS.

Vacuum Seal Off Tip

Inner Envelope

Outer Envelope

Lead Telluride Film (F)
Deposited on
Aquadag Electrodes

Liquid Air

Aquadag
Electrodes (A)
Painted on
Inner Envelope

Infrared Radiation

Cell Window (W)
Thin Hemispherical Pyrex

Tungsten Leads

Cell Electrodes (E)

LEAD TELLURIDE CELL (Scale: 1½)

With these studies in progress, Richard L. Petritz joined the NOL group. He studied the theory of noise in photoconductive cells, seeking to determine the factors which impose an ultimate limit on the sensitivity of cells (Petritz 1956, Petritz 1959, Petritz and Humphrey 1958, Lummis and Petritz 1957). Soon thereafter, James Humphrey joined the group and extended the NOL investigations to PbSe evaporated films (Humphrey and Scanlon 1957, Humphrey and Petritz 1957a and 1957b). Next, R. F. Brebrick was hired to apply his knowledge of physical chemistry to the problem of crystal preparation (Brebrick and Gubner 1958, Brebrick, et al 1959). The NOL team soon worked out some of the details on the phase system for PbS which enabled them to control the electrical properties of the crystals through a vapor-crystal thermal treatment.

The NOL scientists proposed a theory of photoconductivity based upon data obtained from these studies. It was assumed that incident radiation on the PbTe film excites electrons from a filled band to a conduction band. This leaves a hole which also has mobility, although less than that of the electron. However, unless the electron and hole can be immediately separated, there is a high probability of recombination and, hence, no current. If, however, the film contains several p-n junctions, there will be local fields sufficient to separate the electron-hole pairs.

It was suggested that the sensitization process leads to
the production of a large number of p-n junctions. Lead salt
films show micro crystalline structure. A fresh deposit of
PbTe is an n-type semiconductor. The diffusion of oxygen into
the crystalline lattice makes crystallites p-type. The mechanism
proposed agreed qualitatively with observed results.

Since the film structure of the cells that had been pro-
duced were apparently quite complicated, it was decided to con-
tinue the studies on single crystals. Facilities for growing
crystals and for making Hall effect and resistivity measurements
were built. Very little of the fundamental nature was known
about the properties of the lead salts; even the energy gap was
not established.

The width of the forbidden energy gap in a semiconductor is
one of the important parameters used in describing its character-
istics. Even in 1953, Scanlon reported that others had calculated
such divergent values as 0.3 and 1.17 ev (Scanlon 1953). By
using the crystals grown at NOL, he found significant changes in
stoichiometry for PbS as a function of temperature. This deter-
mination provided a significant step toward understanding the
semiconductive behavior of lead salts. The band gap was determined
from Hall effect data at temperatures less than $500^{\circ}K$ as
0.37 ± 0.01 ev.

Measurements of the optical absorption edge in a semiconductor is of interest since it provides information on the forbidden energy gap and can be used to calculate the radiative recombination lifetime of electrons and holes. However, such measurements are extremely difficult to make since PbS, for instance, has an absorption coefficient of approximately 10^4 or 10^5 cm^{-1} at low wavelengths. By using crystals grown in his laboratory, Scanlon was able to grind and polish these to a thickness less than 0.1 mm (Scanlon 1958). These, of course, were very fragile and broke into small pieces. However, the NOL group developed a microscope technique to obtain optical transmission studies on crystal areas as small as about 600 x 50 microns. The spectral absorption measurements agreed well with the reflectance measurements of others at short wavelengths and with transmission data of others at long wavelengths (where the transmittance is highest). However, Scanlon's data added new precision to the determination of the absorption edge. With this measurement, the radiation recombination lifetime was established at 63 microseconds.

The determination of the optical absorption edge marked another significant step at NOL in establishing a useful understanding of the semiconductor behavior of lead salts. The next comparable step was the development of a technique to control the

material concentrations in the lead salt crystals. This was done by heat treatment in a controlled atmosphere (Scanlon 1959) and permitted a detailed study of the band structure.

In less than a decade, the NOL scientists were able to derive an acceptable theory for the semiconductor behavior of lead salts. Their contributions have been of particular value to understanding the physics of materials. It has been an interdisciplinary effort, involving the team work of many scientists.

6

THE SYRACUSE EFFORT

The interest of the military in developing improved infrared sensitive detectors has been described. Paul Ovrebo, as has been noted, had been in charge of tests of the Kiel IV at Wright Field, Ohio. He, naturally, felt the need for improved detectors, and did his best to encourage groups to undertake research toward that objective. While attending the January 1946 meeting of the American Physical Society he described the need for detector research to W. R. Frederickson, chairman of the Physics Department at Syracuse University. This discussion resulted in an unsolicited proposal to Wright Field from Woodrow Johnson of the Syracuse physics staff. In September 1946, a contract was let to Syracuse. The effort was largely directed toward investigations of photo-emissive effects.

Within a year, Johnson left Syracuse and the work was continued by a recent addition to the Syracuse staff, Henry Levinstein. In February 1948, Levinstein redirected the investigations toward infrared photoconductive cells.

After surveying the field of infrared detector development, Levinstein concluded that PbS cell development was in good hands, and PbSe and PbTe offerred the more promising new paths. The latter seemed better suited for experimental investigations.

Support for this work, it must be noted, came from the military with a desire to create improved infrared detectors. The contract monitor, Niel F. Beardsley, (Krasno 1961) worked in close liaison with Levinstein to assure that that goal would be met. However, the Syracuse effort was directed toward understanding the mechanisms of photoconductivity, and thereby aimed to improved cell production.

By 1949, a number of good cells were being made. Since these were almost unique in the U.S.A., there was a large demand for cells by the military, spectroscopists, and others. By 1950, Levinstein had a recipe sufficiently understood so that his undergraduate students could begin their apprenticeship by making cells to meet these demands. When a student had progressed to a point where he was ready to undertake a thesis, he adjusted readily to the preparation of the films required for his project. This, thus, was an effective educational tool as well as a means for meeting the demands for infrared sensitive cells.

Although in this period (about 1950), Syracuse became an important source of infrared detectors, the main interest remained in the research inquiring into their photoconductive properties. That research soon emphasized the characteristics of crystals. Since this is beyond the scope of these essays, that phase of the Syracuse program will not be described. Suffice

it to say that Syracuse rapidly developed as a center for train-
ing scientists in photoconductivity. The research effort also
naturally encompassed nearly all types of photoconductive cells
(Beyen, et al 1959, Levinstein 1959, Levinstein 1962, Levinstein
1965).

Levinstein, through his teaching, writings, and lectures
has had a profound influence on photoconductor research. One is
apt to encounter his former students wherever infrared activity
is undertaken today.

7

THE ATLANTIC CITY CONFERENCE

Within a decade of Cashman's initial success with PbS photo-conductive cells (1944), a rather large number of investigators were engaged in studies of photoconductivity. Cells were readily available from a variety of suppliers. Reasonable descriptions of the photoconductive mechanism were voiced. Research was being extended to materials other than the lead salts - research that was soon to come to important fruition.

The magnitude of this research effort pointed to a need for a conference in which the various workers could exchange their point of view. Late in 1953 plans for such a conference were initiated, and on 4 - 6 November 1954 it was held at the Senator Hotel in Atlantic City, New Jersey. The proceedings of this conference marked the first extended treatment of the subject (Breckenridge, et al 1955).

In the first paper of the meeting, it was noted (Rose 1955) that although photoconductivity had been observed for well over a half century, it was only within the past decade that experiments had been designed to enable one to present data in terms of the lifetime of a free carrier or the capture cross section of bound states. He contended that the lifetime of a free

carrier is the characteristic parameter of a photoconductor. This is so because the photoelectron current is equal to the product of absorbed photon current and the ratio of lifetime to transit time for a free carrier. Rose described the process of termination of the lifetime of a free carrier, noting it to usually be by recombination with a deep-lying bound state in the forbidden zone. This is structure sensitive and, because of its complexity, not amenable to exact solution. He was able to provide some insight into the situation, however, through consideration of deep-lying states, or ground states, and shallow-lying bound states, or traps. The number of ground states increases with increasing radiation density.

A second paper on the phenomenological theory of photoconductivity applied the recently developed concept of information theory to considerations of lifetime and the limit of sensitivity of photoconductors (Petritz 1955). Here, it was concluded that the limit of sensitivity of an intrinsic semiconductor (i.e., where the signal response is equal to noise) is established by fluctuations in the number of conduction electrons and holes, which arise through fluctuations in the rate of emission and absorption of photons and phonons. The ultimate radiation limit can be reached by cooling the lattice. In principle, if the lattice recombination time exceeds the

radiation lifetime one can reach the same limit as that of the cooled detector.

Petritz divided the application of radiation detectors into broad classes. In the first, one deals with weak signals. At the opposite extreme one deals with strong signals. The information rate at these extremes was shown to have a different dependency on the signal to noise ratio. In applications where one's objective is merely to detect the presence of a signal, Petritz showed that an uncooled detector, operated by making the lattice recombination time longer than the radiative lifetime, obtains information at the same rate as an uncooled fast detector whose noise is set by the lattice. Petritz was able to provide a basis for an empirically derived relation of McAlister (of Eastman Kodak Company) which stated that the product of the limit of sensitivity and time constant of a detector is a constant.

Petritz's approach to the photoconductive mechanism and to noise consideration did much to improve the understanding of the optimum use of infrared sensitive detectors.

Following these discussions of the phenomenological aspects of photoconductivity, several investigators described photon absorption processes. This helped clarify the concepts of traps, lattice vibrations, indirect transitions, and other characteristics

associated with absorption. The next topic covered was electron processes, which were discussed in six papers.

Photoconducting materials were next considered. By this time, some attention could be paid to materials other than the lead salts. However, from the viewpoint of this essay the critical paper was that presented by Mahlman, Nottingham, and Slater on the photoconductivity of PbS. This paper reported the results of the research undertaken at the Electronics Corporation of America (which had been Photoswitch, Incorporated until shortly before the conference).

This paper presented the rather extensive experimental measurements of PbS films as well as a theoretical interpretation of those measurements. The measurements were principally of four kinds: the dark conductivity of cells as a function of temperature, the conductivity under different levels of irradiance, the time constant, and the spectral response. By plotting the log of the dark conductivity for a large number of cells as a function of 1/T, a pattern emerged. The curves seemed to evolve into a single straight line at higher temperatures. This was called the Slater line. The slope of this line was interpreted in terms of the forbidden energy gap and indicated a value of 0.35 ev. This value was in agreement with values deduced by other workers from a variety of evidence. From this, the ECA scientists were led to suggest that the photoconductive mechanism in PbS could be

interpreted in terms of a barrier-modulation theory. Such a theory implies that the micro-crystalline structure of the film consists of alternate n and p type elements. If an electron is excited by a photon from the ground state to the conduction band in a p-type element, the resulting hole is bound by the adjacent n-type elements. Conversely, an electron excited in an n-type element becomes bound in the conduction band. This theory suggested how the cell conductivity and time constant will vary upon illumination. The ECA observations gave support to the theory.

The ECA effort was undertaken in the desire to perfect an infrared mosaic tube. Although the tube itself has yet to be perfected, the effort did permit an improved understanding of the photoconductive properties of PbS. The complementary program of NOL has already been discussed (Sec. 5). This effort on the chemical and physical properties of PbS single crystals was described at the Photoconductivity Conference (Scanlon, Brebrick, and Petritz 1956).

From measurements of the Hall effect, the NOL scientists deduced the energy gap to be 0.37 ev. Other measurements indicated a somewhat lower value, but this could be explained by the variation in stoichometry in PbS as a function of temperature. A companion paper (Woods 1956) also presented at the conference suggested that "... the change in resistance (of PbS films) under illumination

occurs principally through a change in the number of carriers in the crystallites, current-amplification effects due to barrier modulation being small."

The theory evolving from studies of lead salts thus showed subtle, but important, differences. This indicates the real value of such a conference. The paths required for further research were delineated.

The studies of the properties of PbS would be understood by considerations of their uniqueness among the lead salts: PbS, PbTe, and PbSe. Moreover, by comparing the properties of these cells with those of intrinsic detectors and doped crystals (principally germanium), the understanding of the photoconductive mechanism would be enhanced.

Somewhat prophetic of these efforts was the paper presented by Levinstein reviewing his investigations of the electrical and optical properties of PbTe films. He reviewed the detailed experimental investigation he had undertaken: dark conductance as a function of reciprocal temperature with oxygen exposure as a parameter, the thermoelectric power as a function of oxygen exposure, the effect of substrate temperature during deposition on the film's ability to accept oxygen, the spectral transmission of a film at different temperatures and degrees of oxygenation, the spectral index of refraction at room temperature and for

heated and cooled samples, the spectral absorption coefficient for various crystals and films, the spectral extinction coefficient and spectral response, the variation of film sensitivity as a function of exposure to oxygen, the variation in spectral response with exposure to oxygen, and the effect of strong radiation on film resistance (for both n- and p-type films).

At this stage, the enunciation of a quantitative model describing all these observations was still beyond grasp. Levinstein, however, did link his observations qualitatively. From the observed absorption coefficient as well as the spectral response, he deduced an energy gap of about 0.25 ev. He envisaged both free lead and tellurium atoms existing as impurities. The former, having energy levels located close to the conduction band, supply conduction electrons. The addition of oxygen provides acceptors, turning the crystallites to p-types. Levinstein was able to correlate his observations and prepare for the formulation of a more quantitative model.

The Atlantic City conference provided considerable catalysis to understanding photoconductivity. It provided an excellant opportunity for cross fertilization in a new and rapidly expanding facet of science. Although there were some important contributions from the British, the meeting was largely American.

Seven years later, a second conference on photoconductivity

was held (Levinstein 1962). This conference attracted a truly international participation, and the range of subjects was much broader. Investigations in photoconductivity continue. These have not only provided infrared sensitive detectors useful in a broad variety of applications, but also the studies of the properties of the photoconductive mechanism have been responsible for an improved understanding of matter.

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DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author)	2a. REPORT SECURITY CLASSIFICATION
University of Michigan Box 618, I.S.T. Ann Arbor, Michigan 48107	UNCLASSIFIED
	2b. GROUP

3. REPORT TITLE

SOME EARLY LEAD SALT DETECTOR DEVELOPMENTS

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

Scientific   Final

5. AUTHOR(S) (First name, middle initial, last name)

D. J. Lovell

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
1968	83	

8a. CONTRACT OR GRANT NO. F44620-67-C-0051	9a. ORIGINATOR'S REPORT NUMBER(S)
b. PROJECT NO. 9769-02	8600-3-J
c. 61445014	9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)
d. 681304	AFOSR 68-0264

10. DISTRIBUTION STATEMENT

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11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY
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      This report describes the history of the early research and development that that was accomplished in the area of lead salt cells, a device for detecting the presence of infrared radiation. The time span covered is World War II and the period immediately following. The efforts of industries, universities, and foreign countries are discussed; however, the pioneering work of Robert J. Cashman is emphasized. The report is a case history of the development of a science into technology showing some of the mechanisms by which science is advanced. Included in the report is a comprehensive bibliography of the subject.

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KEY WORDS	LINK A		LINK B		LINK C	
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Science History						
Lead Salt Cells						
Lead Sulfide						
Infrared						
Photoconductive						